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ISOTOPE MIXES, CORRESPONDING NUCLEAR
PROPERTIES AND REACTOR DESIGN IMPLICATIONS
OF NATURALLY OCCURRING LEAD SOURCES

by

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June 2013

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Lead-cooled Fast Reactors (LFRs) offer great potential for future compact nuclear power systems. The Small, Secure, Transportable, Autonomous Reactor (SSTAR) is a concept for an advanced fast reactor cooled by lead. Such reactors could be improved by using lead that is enriched in radiogenic lead (e.g., ^{208}Pb) in contrast to the average natural isotopic concentration. This improvement is due to the improved neutron reflection and lower neutron absorption cross-sections of the radiogenic isotopes. Artificial isotope separation of lead is cost-prohibitive; however, a natural lead source that is high in ^{208}Pb and low in ^{204}Pb could be used to improve the design of the reactor. The natural variation of lead isotopic content is geochemically investigated to determine if there are favorable naturally occurring lead sources. The results of the investigation are then used in Monte-Carlo simulations with the MCNP5 code to determine the potential benefits of using such a lead composition to the design of a simplified SSTAR-type reactor. The results demonstrate that natural lead sources high in ^{208}Pb could lead to a reduction in the required core enrichment of up to 1 percent; this benefit could also be applied to make the design smaller, or to increase the power output.

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iii

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ABSTRACT

Lead-cooled Fast Reactors (LFRs) offer great potential for future compact nuclear power systems. The Small, Secure, Transportable, Autonomous Reactor (SSTAR) is a concept for an advanced fast reactor cooled by lead. Such reactors could be improved by using lead that is enriched in radiogenic lead (e.g., ^{208}Pb) in contrast to the average natural isotopic concentration. This improvement is due to the improved neutron reflection and lower neutron absorption cross-sections of the radiogenic isotopes. Artificial isotope separation of lead is cost-prohibitive; however, a natural lead source that is high in ^{208}Pb and low in ^{204}Pb could be used to improve the design of the reactor. The natural variation of lead isotopic content is geochemically investigated to determine if there are favorable naturally occurring lead sources. The results of the investigation are then used in Monte-Carlo simulations with the MCNP5 code to determine the potential benefits of using such a lead composition to the design of a simplified SSTAR-type reactor. The results demonstrate that natural lead sources high in ^{208}Pb could lead to a reduction in the required core enrichment of up to 1 percent; this benefit could also be applied to make the design smaller, or to increase the power output.

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TABLE OF CONTENTS

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LIST OF FIGURES

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LIST OF TABLES

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LIST OF ACRONYMS AND ABBREVIATIONS

BWR…………….....…….. Boiling Water Reactor

EBR-1……………….…… Experimental Breeder Reactor

LBE………………….…... Lead-Bismuth Eutectic

LFR ……………….….….. Lead-cooled Fast Reactor

MCNP………………..….. Monte Carlo N-Particle Transport Code

PWR…………….....…….. Pressurized Water Reactor

SSTAR……………….….. Small, Secure, Transportable, Autonomous Reactor

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I.   INTRODUCTION

Recent work to develop a new generation of advanced nuclear reactors, commonly referred to as "Generation IV reactors," has centered on six possible approaches based mainly on various combinations of neutron energy spectrum (i.e., fast versus thermal) and the material selected as the coolant (i.e., water, helium, molten salt, sodium or lead) [1]. One of the promising Generation IV systems, suitable especially for small, compact nuclear power systems, is the Lead-cooled Fast Reactor (LFR), a fast-spectrum reactor concept in which the coolant is molten lead or a related alloy mixture of elements including lead (such as the alloy mixture of lead and bismuth). In particular, the small reactor known as the Small, Secure, Transportable, Autonomous Reactor (SSTAR) [2] uses pure molten lead as its primary coolant.

In the design of LFRs, it is normally assumed that the lead coolant is composed of the average naturally occurring mix of stable isotopes of lead of which there are four: ^{204}Pb, ^{206}Pb, ^{207}Pb and ^{208}Pb. A review of the properties of these four stable isotopes reveals that they vary significantly in terms of nuclear properties that are important to the design of a reactor (e.g., neutron absorption cross-sections at the neutron energies anticipated in the fast reactor design) [3]. In particular, ^{208}Pb (which is the stable end product of the ^{232}Th decay chain) has the most favorable nuclear properties, while ^{204}Pb (which is the primordial lead isotope) has the least.

It is likely that obtaining pure ^{208}Pb from natural lead sources by artificial isotope enrichment processes would be prohibitively expensive. Nevertheless, it is well known that the isotopic abundances of natural sources of lead are variable, and a source of lead with a favorable isotopic mix may well be obtainable in certain natural geochemical settings. The first objective of this thesis is to evaluate this natural variability of stable lead isotopes and to characterize the most favorable isotope mixes that would be available in nature.

It is reasonable to expect that the design and operation of reactors cooled by lead could, in theory, be improved by using an isotopic mix of lead that is higher in ^{208}Pb

since it is an effective neutron reflector and has a lower rate of parasitic absorption than the other stable isotopes of lead. However, the full significance of using ^{208}Pb (or mixtures of isotopes relatively enhanced in ^{208}Pb while depleted in the less favorable isotopes) has yet to be determined. Modern computational methods have the capability to provide answers to this question, and this is the second objective of this thesis.

Therefore, this thesis seeks to answer two questions: (1) what naturally-occurring favorable mixes of lead are available? and (2) what level of design benefit would the use of such favorable isotopic mixes of lead provide?

In order to answer these questions, a detailed literature study and geochemistry analysis of lead isotope sources was used to identify and quantify potential favorable naturally occurring lead sources; and the MCNP5 code [4] was used to analyze a simplified lead-cooled fast reactor configuration to determine the potential design advantages that would result from the use of the various lead isotope mixes. The isotope abundances of the favorable natural sources were used in simulations of a simplified reactor configuration, and the results of these simulations were compared to results using the assumed average natural isotopic composition of lead in order to determine the possible advantages of using mixes high in ^{208}Pb as the primary coolant in an LFR such as SSTAR.

II.   BACKROUND AND MOTIVATION

A.   INTRODUCTION

In 2011, CNN reported "Global energy use is expected to jump 53% by 2035, largely driven by strong demand from places like India and China" [5]. This jump in energy use is coming largely from the developing world, where many of the countries lack the infrastructure or finances to invest in renewable or large-scale nuclear energy. Such countries are thus forced to utilize fossil fuels, which can be more expensive and are environmentally damaging. Nations with advanced nuclear technology are investigating the feasibility of small-scale affordable nuclear reactors that could be developed for use in developing countries where their use could be scaled up as the demand grows in order to answer market demands for power.

One such option that has been investigated in the U.S. is the "Small, Secure Transportable, Autonomous Reactor," or SSTAR, which utilizes molten lead as a coolant in order to provide compactness and enhanced safety while maintaining the security of fissile material [6].

In the development of reactor systems such as SSTAR, it is important that all design options be considered in order to maximize the efficiency and safety of the reactor. Using a more favorable isotopic content of the lead as the coolant in a reactor such as SSTAR could offer design advantages due to the better neutron absorption characteristics. Furthermore if a favorable isotopic content is used, the reactor could be made smaller and more compact or, alternatively, could be operated with a lower enrichment levels in the fuel. In order to maximize the capabilities of lead-cooled reactors such as SSTAR the potential for various lead sources are analyzed in this thesis. Specifically, the effect of isotopic variation on the fuel enrichment level are evaluated, and naturally occurring lead sources are investigated to determine the possibility of obtaining lead sources containing favorable isotopes.

B.   WHERE DOES LEAD COME FROM

It is well understood that stable lead isotopes come from two sources: some lead is primordial (i.e., the direct result of nucleosynthetic processes as discussed below), and some lead is radiogenic (i.e., the result of the radioactive decay of other heavier elements that are radioactive, and also of primordial origin).

About one second after the start of the Big Bang "the stable, light nuclei have formed (^{2}H, ^{3}He, ^{7}Li). " [7] These light nuclei clustered into stars over time and began fusion in a process referred to as stellar nucleosynthesis, resulting in the creation of larger nuclei. This process includes the various stages of ^{4}He burning, then ^{12}C and ^{16}O burning, and finally silicon burning, and "The end products of chains of such reactions are the mass-56 nuclei (^{56}Ni, ^{56}Co, ^{56}Fe). At this point there is no longer energy released in the capture reactions, and the process is halted" [7].

In order to form elements above mass-56, "neutron capture is the primary production mechanism." [7] The process begins when the mass-56 elements undergo "a sequence of neutron-capture reactions…[t]he next step in the process depends on the intensity of the neutron flux." [7] If the neutron flux is low enough to allow the mass-59 nuclei to beta-decay before capturing another neutron, a process called the slow or s-process occurs in stars. "The s-process proceeds in zigzag fashion through most of the stable isotopes, terminating at ^{209}Bi." [7] If the neutron flux is high enough that the nucleus can capture another neutron before decaying, a different process known as the rapid or r-process can take place; this is thought to occur in supernovae and neutron star collisions [8]. "The r-process…can continue until the fission half-lives are as short as the r-process capture times." [7] Following these processes of neutron capture and decay, the heavy elements are formed (see Figure 1). Among these is the primordial stable isotope of lead and the higher radioactive elements destined to decay to the radiogenic stable isotopes of lead.
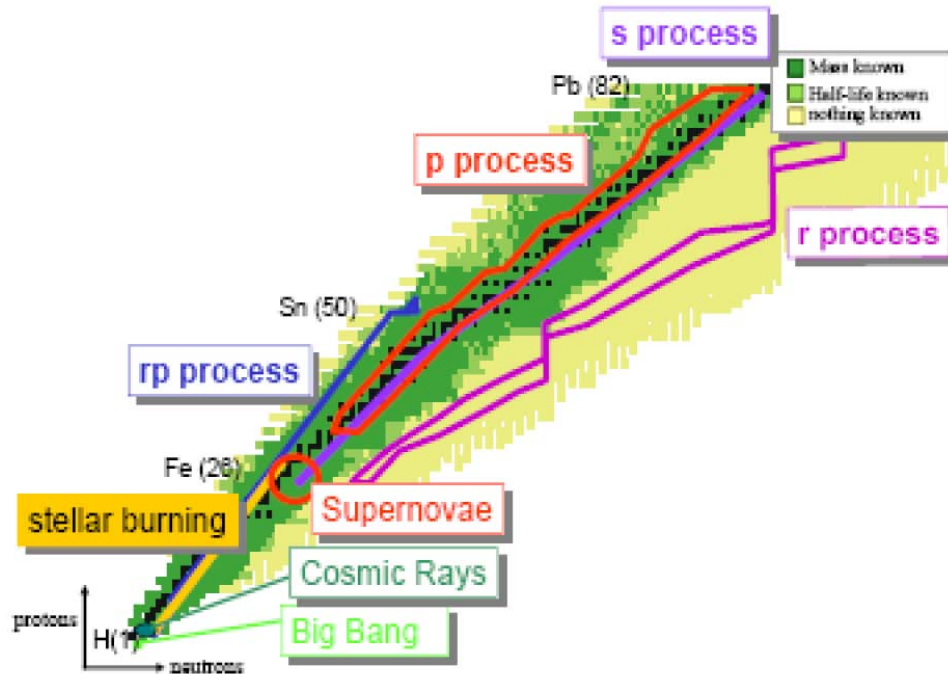
Figure 1.   The chart of nuclides showing the various element creation paths. Lighter colors are less stable while darker ones represent stable elements. From [9].

There are four stable isotopes of lead: ^{204}Pb, ^{206}Pb, ^{207}Pb, and ^{208}Pb [3]. Though all of these isotopes could be created primordially, ^{204}Pb is the only one that must be created primordially. The other isotopes are at the end of three lengthy decay chains. "The nuclides generated during the decay of the very long-lived natural radionuclides U-238 (half-life 4.5 billion years), U-235 (half-life 0.7 billion years) and Th-232 (half-life 14 billion years) are in turn radioactive, and therefore decay again. Thus, the so-called decay chains are created which end only when a non-radioactive nuclide is formed. The uranium-radium decay chain starts from U-238 and ends via 18 intermediate states at the stable lead-206. Uranium-235 is at the beginning of the uranium-actinium decay chain leading via 15 radionuclides to lead-207. With ten intermediate states, the thorium decay chain starting with thorium-232 and ending at lead-208 is the shortest." [10] The three decay chains are illustrated in Figure 2.
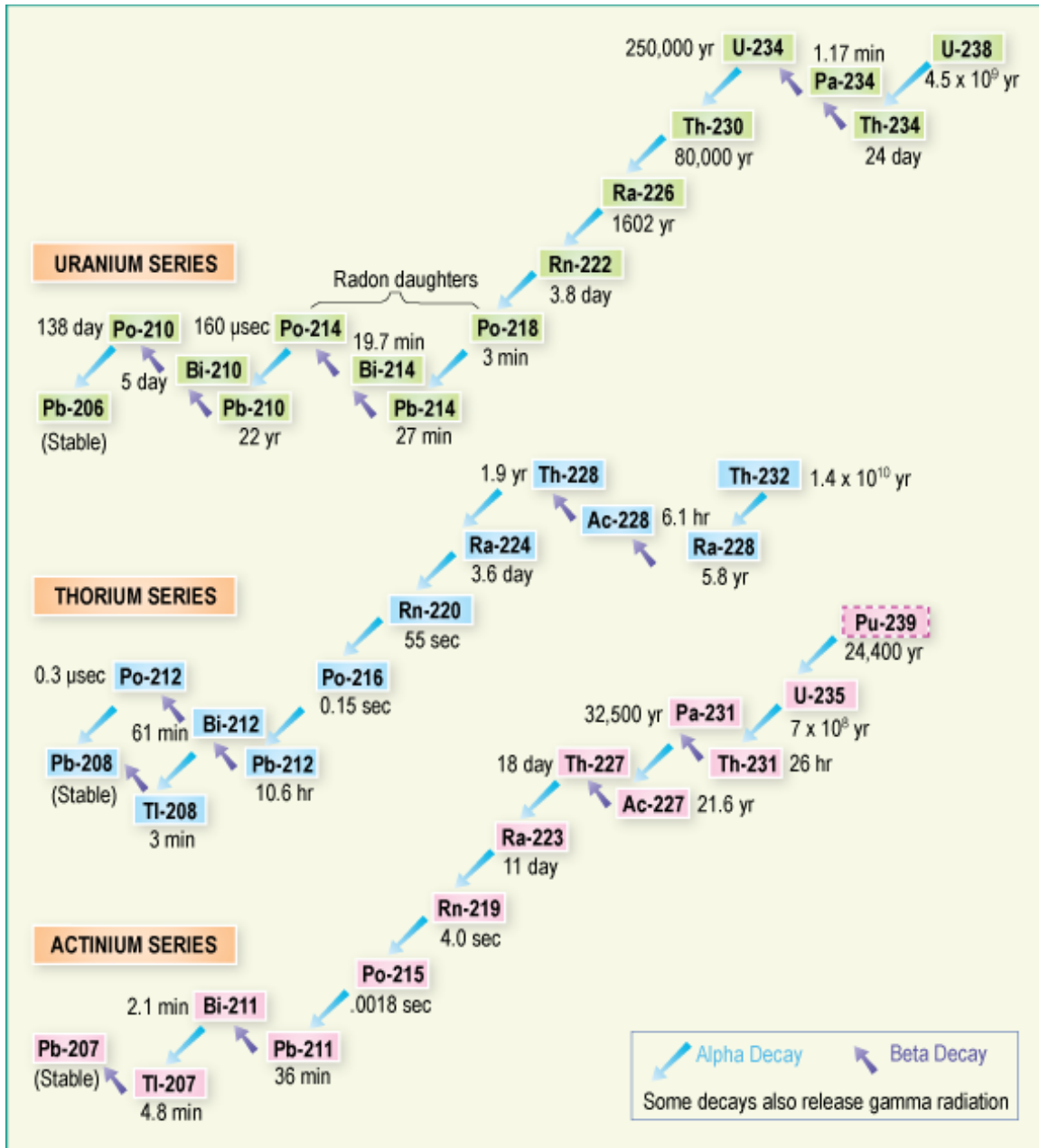
Figure 2. ^{238}U, ^{235}U, and ^{232}Th decay chains resulting in radioisotopes of lead. From [11].

Thus, stable lead comes from both primordial direct creation and as the result of creation of heavier radioactive nuclei that subsequently decay into lead. In natural conditions, the average relative abundance of the four lead isotopes is ^{204}Pb(1.4%), ^{206}Pb(24.1%), ^{207}Pb(22.1%), and ^{208}Pb(52.4%) [3], with all other lead isotopes existing only in trace amounts due to their radioactive decay and relatively short half-lives.

6

C.   NUCLEAR DATA OF LEAD ISOTOPES

To reiterate, the reported natural isotopic composition of lead isotopes is ^{204}Pb (1.4%), ^{206}Pb (24.1%), ^{207}Pb (22.1%), and ^{208}Pb (52.4%) [3]. However, isotope compositions are known to vary considerably depending on the individual sources of lead, even within single geographic areas [12]. In a fast spectrum lead cooled reactor, it is desirable to maximize the reflection of neutrons back into the core, minimize the parasitic absorption of neutrons, and minimize the degradation of neutron energies, thus enabling a compact and hard spectrum (i.e., high average neutron energy) reactor system. Because of the high atomic mass of lead, elastic scattering with any of the isotopes results in high maintenance of the fast energy spectrum of the neutrons. In order to optimize reflection of neutrons and reduce parasitic absorption, the mixture of isotopes in the coolant should have the smallest possible neutron capture cross-section. Of the stable lead isotopes, ^{208}Pb has the lowest capture cross-sections across the energy spectrum, and these absorption cross-sections are also lower than those of the LBE (lead-bismuth eutectic) alloy with natural isotopic abundances of both lead and bismuth [13]. LBE is an alloy that was chosen for early use in molten heavy liquid metal cooled reactors due to its relatively low melting point of 124°C. LBE is composed of 55.5% bismuth and 44.5% lead by weight, and has the advantage of having a very small thermal coefficient of expansion. The major drawbacks of LBE are its somewhat higher tendency toward corrosiveness and its tendency to generate high levels of ^{210}Po, a very toxic radioactive activation product that additionally introduces unwanted heat generation in the coolant.

The following graphs (Figures 3 and 4) show the energy-dependent neutron capture probability of ^{208}Pb compared to that of the other stable lead isotopes, and also that of the average natural isotopic compositions of lead and LBE. In a fast reactor, about 20 to 25% of the neutrons are at energies below 50 KeV [13] where the neutron capture probability is three to four orders of magnitude lower for ^{208}Pb in comparison to the average natural mix of lead or LBE. At higher neutron energies, the margin is lower but ^{208}Pb still enjoys an advantage over the other isotopes and natural mixes.
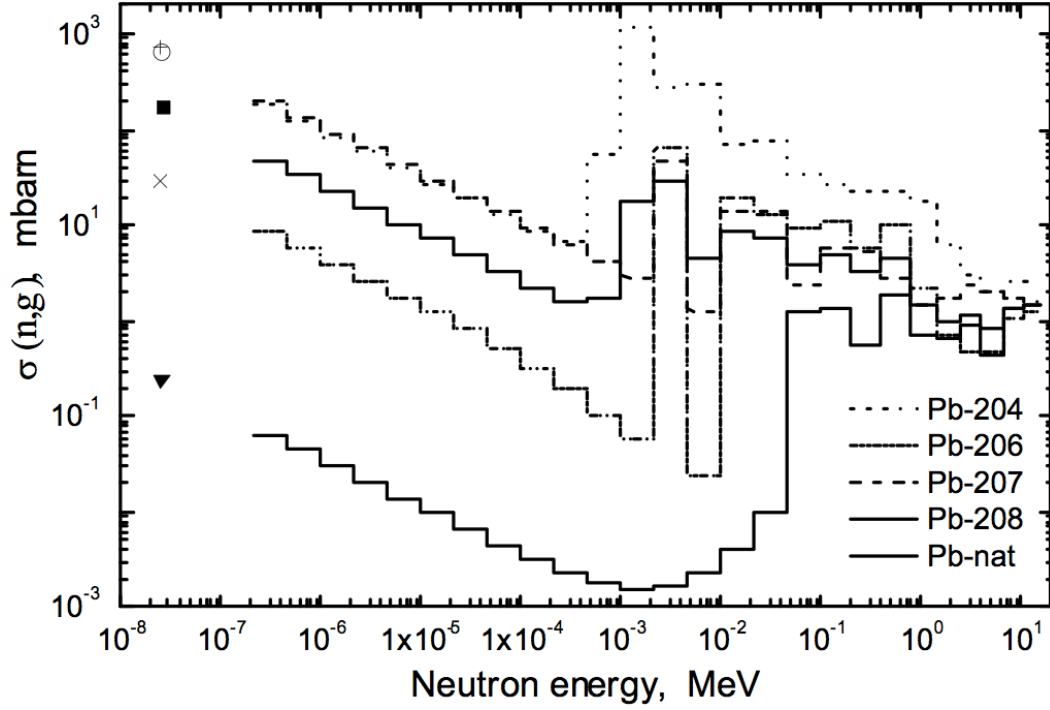
Figure 3.   Cross-sections for neutron capture σ(n,g) in mbarn by stable lead isotopes and by the natural mix of lead isotopes. From [13].
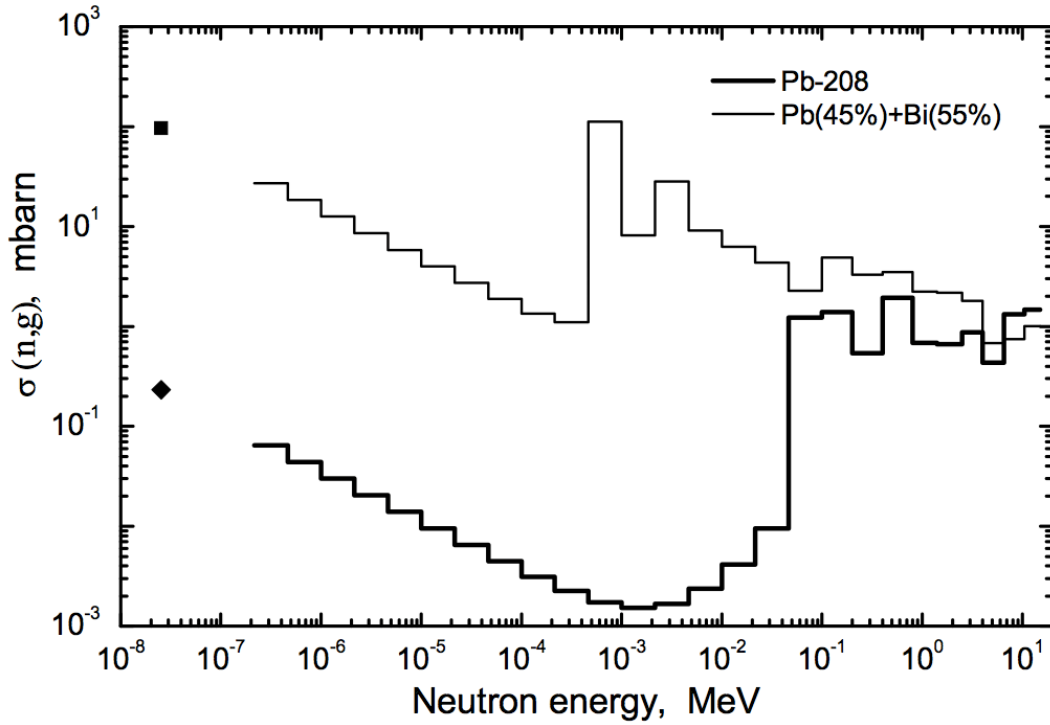
Figure 4.   Cross-sections σ(n, g) in mbarn by stable lead-208 isotope and by the eutectic Pb-nat(45%) – Bi (55%). From [13].

Though nuclear cross-section data is experimentally determined, there are basic principles of physics that determine neutron cross-sections. The two primary determining factors for neutron absorption cross-section are the change in stability of the current nucleus as compared to the nucleus after absorbing a neutron, and whether the nucleus is on the proton rich or the neutron rich side of the valley of stability. The valley of stability is represented by the black (or stable) nuclei in the chart of nuclides (Figure 1), while the neutron rich portion is below the valley of stability, and the proton rich portion is above. There are a number of models that attempt to estimate the stability of a nucleus based upon its number of protons and neutrons; the two primary models are the liquid drop model, and the nuclear shell model [14].

The liquid drop model is based on a formula for determining the binding energy of a neutron by combining five terms: the volume term, the surface term, the coulomb term, the symmetry term, and the pairing term. The volume term accounts for the short range strong force interactions of nearest neighbor nucleons. The surface term accounts for the surface nucleons having fewer neighbors. The coulomb term accounts for the protons' electromagnetic interactions in the nucleus. The symmetry term accounts for the stability added by the neutron-to-proton ratio. And the pairing term accounts for whether the nucleus has an even or odd number of protons and neutrons. The even-even pairings are the most stable, an even-odd arrangement is fairly stable, and an odd-odd arrangement is the least stable. This relationship is due to nucleon pairs being more stable than single nucleons, because the pairs have opposite spins [14].

The shell model is based on the assumption that the nucleus has shells and that complete shells are more stable than incomplete ones, such as is the case with electrons surrounding a nucleus. The evidence for the shell model is first that there are spikes around certain numbers of neutron and protons in the difference between measured and liquid-drop model predicted binding energies. Further, certain neutron and proton numbers have more stable isotopes associated with them than other neutron and proton numbers that match the spikes in measured stability. The spikes in measured stability also align with a drop in the neutron absorption cross-sections. Finally at certain neutron and proton numbers, the energy required to reach excited nuclear states increases. The

neutron and proton numbers are called the magic numbers and they are: for neutrons 20, 28, 50, 82, 126, and for protons 20, 28, 50, 82 [14].

That ^{208}Pb has the lowest neutron absorption cross-section and ^{204}Pb has the largest cross-section makes sense because ^{204}Pb is a proton rich nucleus and ^{208}Pb is neutron rich. It is also relevant to observe the change in stability for each nucleus after absorbing a neutron. Going from ^{204}Pb to ^{205}Pb results in the change from a stable isotope to one that is unstable (i.e., radioactive) with a 17,300,000 year half-life [3] which, though unstable, is a fairly long half life; while going from ^{208}Pb to ^{209}Pb represents a change from a stable isotope to one that is radioactive with a 3.25 hour half-life [3], representing a dramatic loss of stability. Furthermore ^{208}Pb is a double magic number nucleus, while all other lead isotopes have only one magic number. The ^{207}Pb and ^{206}Pb neutron absorption cross-sections vary back and forth at different neutron energies and this also makes sense because, though ^{207}Pb is closer to a magic number, it is an even-odd arrangement, and ^{206}Pb is even-even but is further from a magic number.

Unfortunately the variability in isotopic composition of lead found in nature does not include sources with pure ^{208}Pb, so alternative sources of mixed isotopic composition must be investigated in order to find lead sources with favorable mixes of stable lead isotopes. Enrichment of lead has also been considered but has been found to be prohibitively expensive with estimated costs of "1000–2000 U.S. \$/kg." [13]

D.   NATURAL VARIATION IN LEAD SOURCES

For the purpose of using lead as a coolant in fast reactors, neutron absorption is a very important factor; it is therefore also important that a low concentration of less favorable isotopes and a high concentration of favorable isotopes be sought. Due to the very high cost of artificial lead isotope enrichment, the best mix lead isotopes available in in natural sources should be identified. Even if it is determined that any particular isotopic mix will be acceptable for use in a reactor design, it is also important to know the specific isotope abundances of the actual lead coolant to be sure that the reactor operates according to its design specifications and without any unexpected shifts in the neutron balance. In addition, the exact isotopic composition of the lead coolant must be known

10

during the design phase as the requirements for the other design characteristics such as the parameters related to the coolant circulation and the composition of the fuel will depend on the neutron balance which is strongly affected by the amount of neutron absorption in the coolant.

As early as 1960 it was realized that lead from different environmental/ geochemical sources had variable isotopic makeup. "The isotopic composition of lead varies in lead ores in significant ways. The least abundant isotope, Pb^{204}, is inherited from the original or primeval lead of the solar system; whereas the three most abundant isotopes, Pb^{208}, Pb^{207}, and Pb^{208}, are in part original, in part radiogenic. These three continue to increase in abundance because they are stable end-products of the radioactive decay of uranium and thorium." [12]
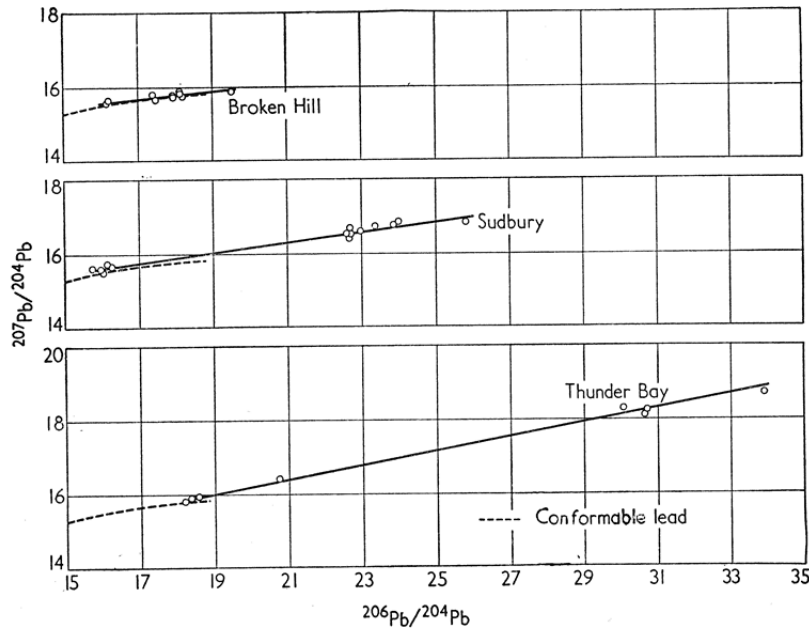
Figure 5.   Chart showing variation in the isotopic content of lead from three different mines, two in Canada and one in Australia. From [15].

Figure 5 gives the $^{207}Pb/^{204}Pb$ ratio on the vertical axis and the $^{206}Pb/^{204}Pb$ ratio on the horizontal. The points show the specific isotope ratios of samples taken from three lead mines, the line being representative of the ratio's progression over time, and the data can be used to establish the age of the particular deposit. For the purposes of the current

research it is primarily of significance that there exists natural variation in the isotopic composition of lead.

Another location where variations in isotopic content of lead have been realized is the Mississippi Valley.
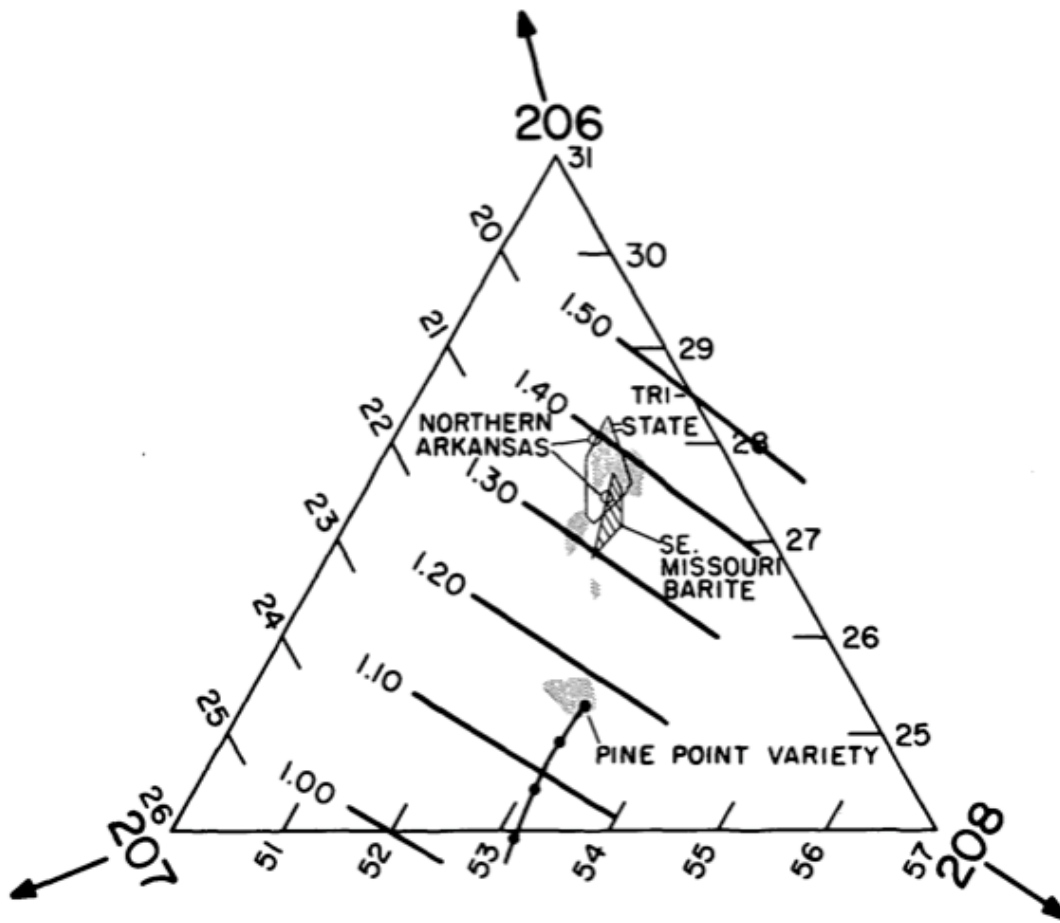
Figure 6.   Graph of the variations of the radiogenic isotopes of lead at various points of the Mississippi Valley. From [12].

Figure 6 presents the concentrations of the four stable radiogenic lead isotopes on one chart with ^{208}Pb abundance being represented along the bottom, ^{207}Pb on the left side, ^{206}Pb on the right side, and ^{204}Pb being represented by the parallel lines through the center. It can be seen in Figure 6 that just in the limited data from the Mississippi Valley there is up to 1% variation in ^{208}Pb, 2% in ^{207}Pb, 3% in ^{206}Pb, and 0.3% in ^{204}Pb. Thus, it

is known that significant natural variation in the isotopic content in lead exists; however, given this fact, if additional and perhaps more diverse (e.g., those not necessarily selected because of their very high total lead content, but rather because of their association with radiogenic precursors) sources are considered, it may be possible to find natural isotopic mixes that are even more favorable than the ones shown in Figure 6.

E.   HISTORICAL DEVELOPMENT OF NUCLEAR POWER

The potential of fissioning uranium for the generation of power was first observed when "Lise Meitner and her nephew Otto Frisch, working under Niels Bohr…calculated the energy release from this (uranium) fission as about 200 million electron volts. Frisch then confirmed this figure experimentally in January 1939." [11] It was later shown by Hahn and Strassman "that fission not only released a lot of energy but that it also released additional neutrons," [11] thus causing additional fissions in other uranium nuclei allowing for the possibility of a self-sustaining chain reaction releasing energy. "This suggestion was soon confirmed experimentally by Joliot and his co-workers in Paris, and Leo Szilard working with Fermi in New York." [11] Shortly after these developments "Bohr…proposed that fission was much more likely to occur in the uranium-235 isotope than in U-238 and that fission would occur more effectively with slow-moving neutrons than with fast neutrons." [11] The benefit of slow moving neutrons is that the fission cross-section of uranium-235 is much greater at lower neutron energies than it is at the fast neutron energy of neutrons as they are released by fission of uranium. This discovery led Szilard and Fermi to suggest the use of a moderator to slow down the emitted high-energy neutrons. With the initial pieces put together, the first artificial nuclear reactor, Chicago Pile #1, was built in the basement at the University of Chicago, and reached criticality on the second of December in 1942 [16].

The Chicago Pile #1 Reactor (CP-1) used graphite as a neutron moderator and natural uranium as fuel, as there was not a process developed yet to enrich uranium in the 235 isotope. After the success of CP-1, much of the emphasis and funding was moved to the development of atomic weapons until the end of World War II. During the effort to produce the nuclear bomb, a process to enrich uranium was engineered, and after the war

many scientists turned their attention to using nuclear energy to meet energy demands. Most of the early work in reactors was focused on thermal reactors because thermal neutrons have a much greater fission cross-section than that of fast neutrons; further, in a thermal reactor the delayed neutrons make the reactor more easily controllable than is the case for a fast reactor. However, the potential of fast reactors being able to efficiently convert ^{238}U into ^{239}Pu (a fissile material) was realized early on and has been researched since.

Figure 7. The first use of nuclear electricity, the lighting of 4 light bulbs in the EBR-1 facility. From [17].

In fact, the first nuclear reactor to successfully produce electricity "was the small Experimental Breeder reactor (EBR-1), a fast reactor designed and operated by Argonne National Laboratory and sited near Idaho Falls, Idaho. The reactor started up in

December 1951." [11] Although the first successful application of nuclear power to produce electricity (see Figure 7) was a fast reactor, most subsequent development of commercial reactors was based on water-cooled thermal reactor technology resulting in the modern high efficiency boiling water (BWR) and pressurized water (PWR) technologies.

F.   FAST REACTORS

An important limiting factor for current thermal reactor technology is the cost and system complexity associated with the fuel cycle including fuel enrichment, fabrication, and the transport and disposal of spent nuclear fuel. This is strongly influenced by the fact that natural uranium is composed of 99.3 percent ^{238}U, and 0.7 percent is ^{235}U, which is the fissile component of uranium. This small concentration of this fissile component and the low enrichment of most commercial reactor fuel reduce the ability to fully extract the energy content of the uranium fuel. The "capability to extract the potential energy in the uranium fuel is limited to less than 1% of that available." [18]

A fast reactor is one that uses high energy or 'fast' neutrons rather than thermal or 'moderated' neutrons. In order to keep the neutrons at high energies, the coolant must not degrade the energy spectrum by allowing significant moderation of the neutrons. Therefore, fast reactor designs avoid the use of light materials in the coolant and structure, and instead rely on heaver elements that maintain high neutron energies following elastic collisions. "Although these fast neutrons are not as good at causing fission, they are readily captured by an isotope of uranium (U238), which then becomes plutonium (Pu239). This plutonium isotope can be reprocessed and used as more reactor fuel." [19] Lead and sodium are potential coolants for use in fast reactors.

The ability to convert the non-fissile (238) isotope of uranium into a fissile isotope of plutonium dramatically increases the energy utilization of the uranium fuel. This process, called "conversion" means that the otherwise "unutilized energy can be harvested, thereby extending by a hundred-fold the amount of energy extracted from the same amount of mined uranium." [18] Under some conditions, the reactor can produce more fissile material than it consumes, a process called breeding, Fast reactors also allow

15

for the reuse of "fuel from thermal reactors and the depleted uranium from the enrichment process." [18]

The operation of fast reactors is largely facilitated by the difference in the cross-sections of ^{235}U and ^{238}U, where the absorption cross section is lower than the fission cross section in ^{235}U as shown in Figure 8. For ^{238}U the absorption cross-section is usually five orders of magnitude larger than the fission cross-section for energies lower than 1Mev as shown in Figure 9, facilitating the reactor's capability of breeding ^{239}Pu.
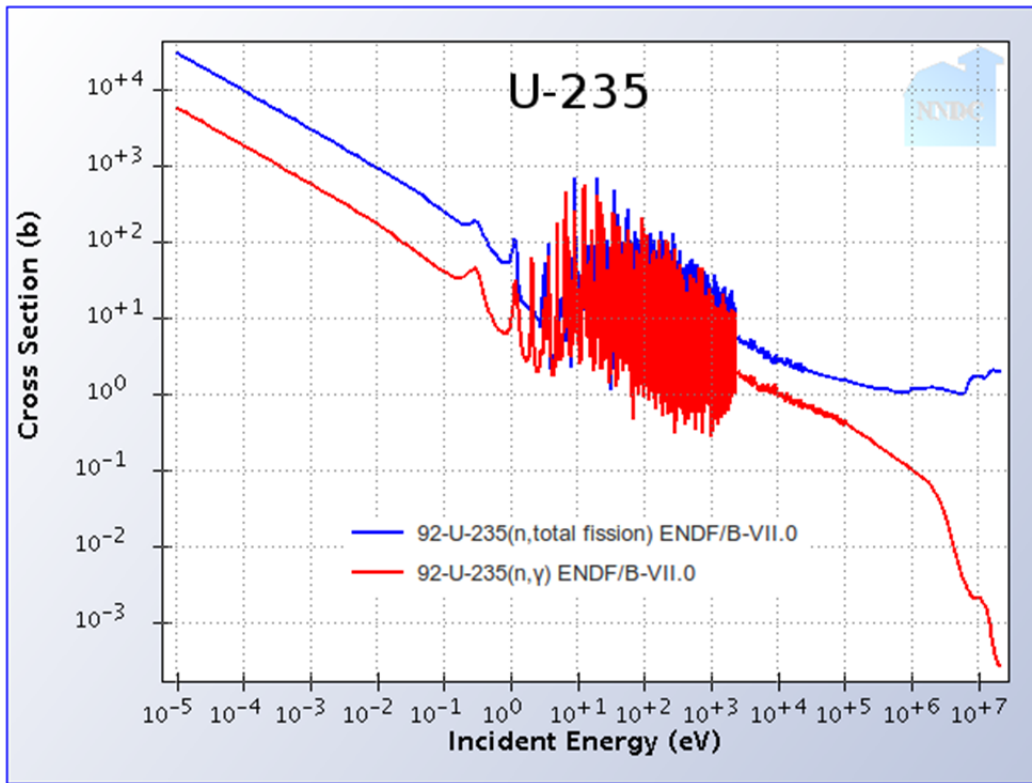
Figure 8.   Comparison of the neutron capture and the fission cross section for U-235. From [20].
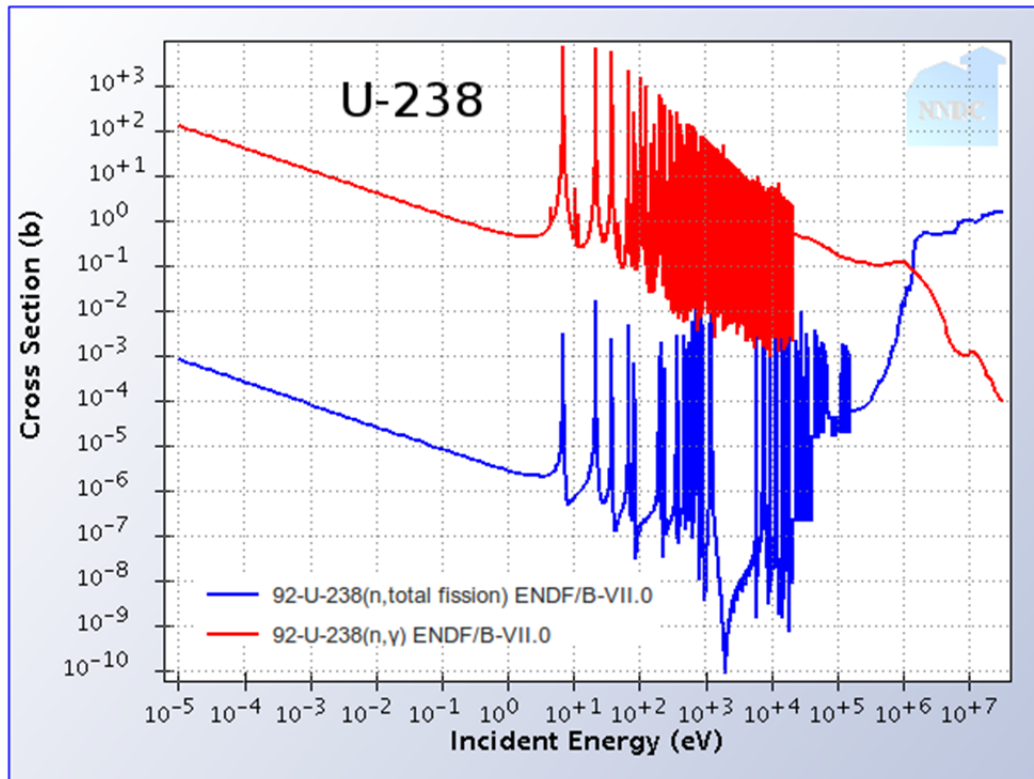
Figure 9. Comparison of the neutron capture and the fission cross section for U-238. From [20].

A secondary effect of fast reactors is that they can greatly lower the amount of radioactive waste produced when combined with fuel recycling. The ability to utilize the ^{239}Pu and other minor actinides created in the reactor means that much of the radioactive waste created in the reactor can be consumed in another reactor [18].

According to the American Nuclear Society:

Fast reactors in conjunction with fuel recycling can diminish the cost and duration of storing and managing reactor waste with an offsetting increase in the fuel cycle cost due to reprocessing and fuel refabrication. Virtually all long-lived heavy elements are eliminated during fast reactor operation, leaving a small amount of fission product waste that requires assured isolation from the environment for less than 500 years. [18]

With the noted benefits of fast reactors there has been and is continuing worldwide interest in developing advanced fast reactor technologies as part of the international Generation IV initiative [1]. Among the promising Generation IV fast

reactor technologies, reactors cooled by lead and sodium (i.e., liquid metal cooled systems) are prominently considered because of the ability of these coolants to effectively cool the reactor core while reflecting rather than absorbing neutrons with a minimal energy loss.

G. LEAD COOLED REACTORS (LFRs)

Liquid metals are commonly considered as advantageous coolants in fast spectrum nuclear reactors because, in contrast to water or other materials with light atomic components, metallic elements (and particularly heavy metallic elements) have a much lower propensity to slow down neutrons in collisions than do lighter (and especially hydrogenous) materials; and, in addition, due to their low vapor pressures and high boiling points, they do not require high operating pressures to maintain their liquid state. Though liquid metals other than lead, such as mercury and liquid sodium, are viable as reactor coolants, they have their own disadvantages. Mercury has the disadvantage of a higher vapor pressure, lower boiling point, and higher toxicity. Sodium carries the disadvantage of high chemical reactivity. Lead has the disadvantage of a relatively high melting point; however it also has the advantage of a very high boiling point. In the previously mentioned Soviet submarine propulsion program, LBE was used as the reactor coolant because it has a substantially lower melting point than does pure lead: LBE melts at 124°C and pure lead melts at 327°C [21]. However, LBE is somewhat more corrosive in contact with steels than pure lead [21]. LBE also has the advantage of having a low coefficient of thermal expansion near the melting point and will experience a long-term expansion upon freezing; however lead will contract upon freezing and, when in the liquid phase will expand significantly with an increase in temperature [21]. The use of pure lead provides an advantage in corrosive properties, however the coefficient of expansion and the contraction when melting present engineering challenges. For the LBE coolant, the presence of bismuth leads to the production of Po-210 upon irradiation and this represents several disadvantages as a result of its high radiotoxicity and the heat load that results from its presence in the LBE coolant. In contrast, pure lead has several advantages over the lead bismuth eutectic as was described in a recent IAEA publication [22] and summarized in Table 1.

18

Pro(+) and contra(-) of pure lead vs LBE		
Melting Point (°C)	Pb has a higher (327 °C) melting point than LBE (125 °C)	-
Melting expansion	Pb has a higher volume increase upon melting	-
Expansion at solid state	Pb does not expand at solid state (~1,2% linear expansion of LBE in one year after solidification and cooling at room temperature)	+
Thermal conductivity	Pb has a higher thermal conductivity (17,7 W/(m*K) than LBE 14,3 W/(m*K)) (data at 500 °C)	+
Slag formation	First tests do not show slag formation in Pb	+
Dust formation	Strongly reduced	+
Corrosion	Pb is less corrosive than LBE at equal temperature	+
Long term Radio-toxicity (inhalation)	Reduced by about four decades	+
Long term Radio-toxicity (ingestion)	Reduced by about three decades	+
Availability	Pb is largely available	+
Cost	Pb is cheap	+
Polonium generation	Reduced by about four decades	+

Table 1.   Chart of the advantages and disadvantages of pure lead over LBE (lead-bismuth eutectic) for use as a reactor coolant. From [22].

Though the higher melting point of pure lead presents a significant design and operational challenge, lead's other properties suggest the possibility of a reactor that is simpler to build and operate than a traditional water-cooled reactor, providing an advantage for use in LFR systems such as SSTAR, described in more detail below. According to L. Cinotti, "It is expected that the properties of lead (expecially?? (sic) high

boiling point and low chemical reactivity with air and water) allow to conceive reactors easy to operate." [22]

Lead (in the form of LBE) was initially used as a coolant in nuclear reactors when the Soviet Union built eight Alpha class submarines with reactors cooled with LBE. The overall program included several land- and sea-based prototypes and overall the program consisted of 12 reactor vessels and 15 cores [23]. The lead-bismuth reactors were selected over water cooled reactors in an effort to provide compactness to the submarine; this, along with the extensive use of titanium, according to the Federation of American Scientists sources, resulted in a 3800 ton submarine capable of 43–45 knots submerged. The first was commissioned in 1972 and "it could accelerate to a speed of 44.7 knots, a record which will hardly be beaten in the near future." [24]

Aside from naval propulsion, an important motivation for the investigation of lead cooled reactors, specifically small, portable models, is to address the accelerating need for clean and efficient power in the developing world. "Most commercial nuclear reactors are large light-water reactors (LWRs) designed to generate 1,000 megawatts electric (MWe) or more. Significant capital investments are required to build these reactors and manage the nuclear fuel cycle. Many developing countries do not need such large increments of electricity. They also do not have the large-scale energy infrastructure required to install conventional nuclear power plants or personnel trained to operate them. These countries could benefit from smaller energy systems, such as SSTAR." [2]

SSTAR or the Small, Secure (or Sealed), Transportable, Autonomous nuclear Reactor is a design that was developed with the support of the U.S. Department of Energy (DOE) in "an initiative to develop a conceptual design of a reactor that will deliver nuclear energy to developing countries and significantly reduce the proliferation concern associated with expanded use of nuclear power. Three national laboratories are collaborating on this initiative. Lawrence Livermore, which leads the collaboration, is researching materials and coolants for the reactor and evaluating how it can be deployed. Argonne is designing the reactor, and Los Alamos is contributing its expertise on coolant and fuel technologies." [2]

The SSTAR could provide many advantages over traditional reactors. First it would be transportable, "the reactor will be about 15 meters tall by 3 meters wide and will not weigh more than 500 tons—small and lightweight enough to be transported on a ship and by a heavy-haul transport truck." [2] Also SSTAR would not require an expansive nuclear-industrial infrastructure. "Because the supplier nation will provide both the reactor and the associated fuel-cycle services, the host nation can produce electricity without needing an independent supply of uranium or other fuel at the front end of the cycle. The host nation also won't have to dispose of the nuclear waste at the back end of the cycle." [2] Furthermore "the current SSTAR design (Figure 10) reduces the potential for a proliferator or terrorist to divert or misuse the nuclear materials and technology. Nuclear fuel will be contained within the sealed, tamper-resistant reactor vessel when it is shipped to its destination, and the spent reactor core will be returned to the supplier for recycling." Finally due to the simplified design with operational autonomy and cost reductions associated with mass production, the cost of electricity should decrease over time.
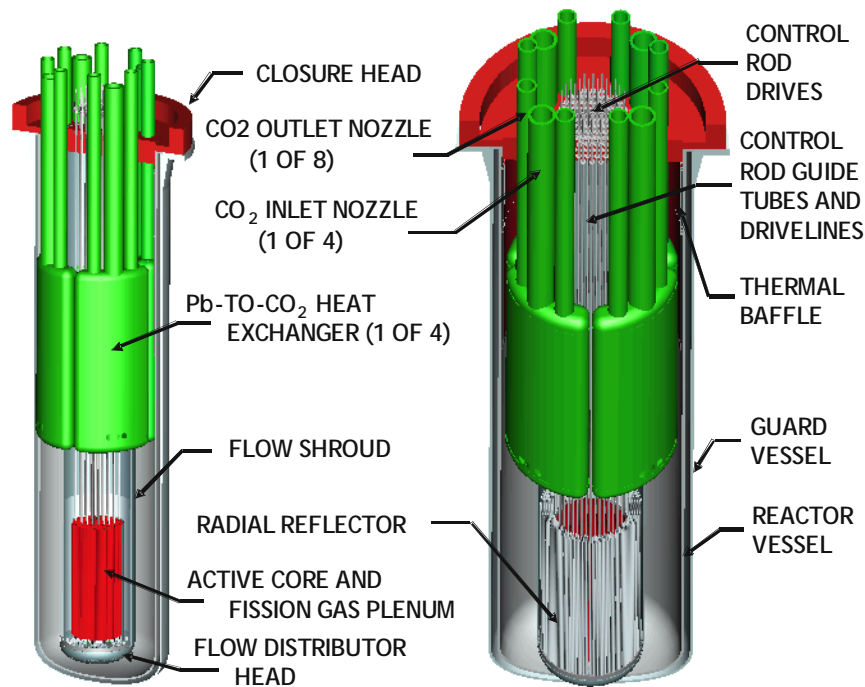
Figure 10. A depiction of the SSTAR design estimated to be roughly 3 meters in diameter and 15 meters tall. From [25].

H. POTENTIAL EFFECTS OF ISOTOPIC COMPOSITION ON REACTOR DESIGN

In the design of an LFR such as SSTAR, the characteristics of the system could be improved by using an isotopic mix of lead that is higher in Pb^{208}, due to the more favorable nature (primarily the lower neutron capture cross-section) of this isotope in comparison to the other stable isotopes of lead. The lower capture cross-section means that more neutrons will be reflected back into the core rather than absorbed by the lead coolant. This will result in greater design flexibility for such systems, allowing either greater compactness in physical dimensions or reduction in the required fissile content of the fuel. The potential design implications are that a reactor unit could use lower nuclear fuel enrichment than an equivalent unit with a standard isotopic mix of lead. Another possible benefit is that the reactor core or potentially the whole unit could be made smaller and thus more portable with the same energy output. The net result of using a lead source that is high in Pb^{208} (and correspondingly reduced in the less favorable isotopes) is that the reactor could be made more portable, able to operate with a lower enrichment fuel, or provide a greater energy output with the same nuclear fuel.

III. THEORY AND METHODOLOGY

The sciences of geochemistry and nuclear physics have been applied in an effort to answer the questions: "(1) What naturally-occurring favorable mixes of lead are available? And (2) What level of design benefit would the use of such favorable isotopic mixes of lead provide?" To determine the available natural mixes of lead geochemical analysis have been performed. In order to model the system, the principles of nuclear physics have been used to develop a sufficiently accurate simplified model of a reactor to answer question 2. The MCNP5 software package, which is based upon Monte Carlo methods, has been used to perform simulations of the reactor model to create meaningful results.

A. GEOCHEMISTRY

Geochemistry is the science of using chemistry to describe and explain geological systems and occurrences. This thesis has investigated lead geochemistry to determine where favorable isotope mixes of lead would likely be located, and how favorable the naturally available mixes are. "Geochemists study the occurrence and distribution of chemical elements in rocks and minerals." [26] Geochemistry includes the "study the movement of these elements into soil and water systems." [26] With information on where relevant ores such as thorium and lead are found in nature and how they were initially created, it is anticipated that favorable isotopic mixes of lead, especially those high in ^{208}Pb, can be found in nature.

"The natural abundance of thorium is ~3× greater than that of uranium, whereas the size, valence, and crystal-chemical properties of Th^{4+} are nearly identical to those of U^{4+}." [27] This commonality between the two elements, suggests similar geochemical transport, however there are a few key differences that lead to variations in the local concentrations of the two elements. The processes that determine uranium and thorium concentrations can be divided into four time phases of geochemical epochs represented by the time period during which they occurred (in units of Ga, or billions of years, before

23

the present time): phase I (~4.5 to 3.5 Ga), phase II (~3.5 to 2.2 Ga), phase III (2.2 Ga), and phase IV (.4 Ga) [27].

During phase I "Uranium and thorium are trace elements, with estimated whole-Earth concentrations of >10 and ~30 ppb…Therefore, before any U or Th minerals can form, the elements must be transported and concentrated locally by many orders of magnitude." [27] The uranium and thorium were collected near the crust of the earth through "low-density, Si-rich melt phases" [27] that were chemically possible due to uranium and thorium being "highly incompatible in all common rock-forming silicates except for zircon ($ZrSiO_4$), which can accommodate several weight percents of the isostructural coffinite ($USiO_4$) or thorite ($ThSiO_4$)." [27] This process led to uranium and thorium "enrichment factors [in the crust] of at least 100 compared with the mantle." [27] The two elements were further concentrated at the crust during phase I via magmatic hydrothermal activity because though "Uraninite is highly insoluble as the U^{4+} species $UO_{2(aq)}$ in aqueous solutions with pH > 3 and at temperatures up to about 300 °C... U^{4+} dissolves readily at low pH (< ~4) in the presence of fluoride that forms soluble U^{4+}-F complexes." [27] The same is true for thorium; furthermore magmatic hydrothermal activity separated the two elements. "Separation of U from Th under these circumstances can be expected to occur if chloride- or CO_2-rich fluids become involved in magmatic processes because complexing of U^{4+} by chloride or carbonate is apparently much stronger than for Th^{4+} at elevated temperatures and pressures." [27] A final method of concentrating uranium and separating uranium from thorium "relies on U in the 6+ oxidation state. If near-surface U^{4+} becomes oxidized, for example through UV photo-oxidation, auto-oxidation, or near-surface oxidative weathering, the resulting uranyl complexes are highly soluble." [27]

During phase II the uranium decay leads to a change of U^{4+} to Pb^{2+} which has the secondary effect of creating the highly soluble, and therefore highly mobile, U^{6+} state [27]. The solubility increase causes further separation of uranium from thorium. In contrast thorium, due to its longer half-life suffers less radiation damage, is less soluble than uranium, and does not have a highly soluble 6+ state, thus is has a much lower mobility [27].

During phase III the uranium in solution was exposed to more oxygen as the atmosphere began during the "great oxidation event," this lead to the creation of various uranium minerals, and during phase IV organic and biological processes furthered the mineralization of uranium [27]. During this time thorium was transported geochemically in the same way that rare earth metals were leading to some greater variation of thorium deposits [28].

Though lead falls out of the uranium and thorium transport processes, the vast majority of lead deposits were created by lead from below the earth's crust that traveled to the crust via volcanic activity. The two primary processes are "sedimentary exhalative deposits," which occur when metal-rich hot liquids come into contact with water. The metals precipitate when they interact with the sediment at the bottom of the water, leading to the creation of ores. Also "volcanogenic massive sulfide deposits" can lead to the creation of lead deposits, these events are formed by the "black smoker" sea vents [29].

The various lead sources from the mantle will likely be the same or similar in isotopic content due to the high levels of mixing in the mantle; the lead that is created by the decay of thorium and uranium will be of greater interest as it is likely to have large variations in isotopic content.

B. NUCLEAR PHYSICS

Nuclear physics analysis is the study of the causes and effects of the fundamental forces in play within the nucleus of atoms. Due to the three most common isotopes of lead being the result of the nuclear decay of other elements, and most geochemical analysis of lead being concerned with the age of ores and deposits as determined from the isotopic and elemental composition, the Bateman, and half-life equations have been used to investigate isotopic concentrations of lead concentration within lead ores based upon calculated ages.

Important methods of nuclear physics used in this thesis include the Bateman equations for calculating the buildup and decay of radioactive and radiogenic isotopes in decay chains; radiological dating methods, and basic nuclear reactor physics.

1. Bateman Equations

The Bateman equations are equations that are used to solve for the activity ($\mathcal{A}$) in the nth member of a decay chain in terms of the decay constants of previous terms and the number density or amount of the parent isotope (N_0) [7].

$$\mathcal{A}_n = N_0 \sum_{i=1}^{n} c_i\, e^{-\lambda_i t}$$

where:

$$c_m = \frac{\prod_{i=1}^{n} \lambda_i}{\prod_{i=1}^{n}{}'(\lambda_i - \lambda_m)}$$

$\mathcal{A}_n$ = daughter activity

N_0 = number density or amount of parent isotope

λ = decay constant

t = time

Equation 1: Bateman equations. From [7]

The Bateman equations use the assumption that the rate of decay of a radioactive isotope is constant throughout time and space in order to determine the activity of a daughter in the decay series from the amount of the parent isotope, taking into account the radioisotopic decay rates and elapsed time.

2. Radiological Dating

Radiological dating is the use of ratios of parent to daughter nuclei to determine the age of a sample. This method can only be completely accurate if there are a significant number of nuclei to provide for statistical accuracy, and the initial and current ratios of the nuclei are accurately known [7]. The equation used for Radiological dating is:

$$\Delta t = \frac{1}{\lambda} \ln \left(1 + \frac{N_D(t_1)}{N_P(t_1)} \right)$$

N_D = number of daughter isotopes

N_P = number of parent isotopes

λ = decay constant

t = time

Equation 2: Radiological dating equations. From [7]

Equation 2 is only valid when the initial daughter concentration is zero; however in the geological time scale, the initial concentration will be assumed to be low enough for equation 2 to give meaningful results. This is a necessary assumption due to the limited data available.

Radiological dating also uses the assumption that the rate of decay of a radioactive isotope is constant throughout time and space in order to age samples with known ratios of daughter and parent isotopes.

3.   Reactor Physics

Reactor physics is a branch of physics applicable to nuclear reactors. The basic principle behind nuclear reactors is fission, which is the splitting of atoms by incident neutrons resulting in a release of energy and generation of additional neutrons. There are three primary fissile materials, the 235 and 233 isotopes of uranium and the 239 isotope of plutonium. A fissile material is one that is capable of readily sustaining a fission chain reaction. A reactor works by assembling a concentration of fissile material that is self-sustaining as the neutrons released by a given generation of fissions incite a similar number of fissions in the next generation, thus creating a stable chain reaction. The fission reactions create heat, which is then transferred by a coolant and finally harnessed by an external power conversion system. The ratio of the number of neutrons in a current generation divided by the number of neutrons in a previous time step is known as the

criticality constant, or k_{eff}, of a reactor. Criticality is an important measure of a reactor as a k_{eff} above 1 will lead to an increase in power and, if not properly controlled can result in accumulation of heat that may ultimately cause overheating and in extreme cases a meltdown; while a k_{eff} below 1 indicates a sub-critical condition which is incapable of sustaining the generation of power. Ideally a reactor will have a K_{eff} of exactly 1 in order to maintain a safe and usable nuclear chain reaction; in practice the value is usually changing but stays near 1 as the system constantly adjusts to changes in its criticality conditions [7].

Another important concept of reactor physics is that of cross-section. Cross-sections are measured in in units of area, but are not representative of actual area; they are instead representative of the probability that a particular type of nuclear interaction will occur. Also, cross-sections of interactions vary with the energy of the reaction. This information is essential in aiding reactor designers to build a system that keeps undesired reactions to a minimum, by using materials that have low cross-sections for undesired reactions, and high cross-sections for desired reactions [7].

C.   MONTE CARLO METHODS

Monte Carlo methods are a class of computational algorithms that utilize random number sampling in order to solve complex problems. The methods are computationally intensive and are therefore used most often on problems with many variables and a large domain of possible inputs, where closed form or deterministic solutions would be impossible. Monte Carlo methods are used primarily in the solution of optimization and numerical integration problems. "Monte Carlo is often the method-of-choice for applications with integration over many dimensions," [30] The applications of the software extend to fields such as "high-energy physics, particle transport, financial analysis, risk analysis, [and] process engineering." [30] The process of a Monte Carlo method is to define an input domain, randomly or pseudo-randomly generate a large number of inputs, calculate the results corresponding to each input set and use the aggregate of the results to determine an overall solution. In order to obtain accurate results the Monte Carlo method must properly simulate the problem and calculate a large

number of iterations. To complete analyses of reactor criticality under varying assumptions of lead coolant isotopic composition, the code MCNP5 was utilized.

D.   MCNP5

MCNP stands for **M**onte **C**arlo **N-P**article Transport Code; it is a code developed and maintained by LANL, Los Alamos National Laboratory, and the '5' denotes that this is the fifth release of the software package. The method was initially developed by Von Neumann, Ulam, Fermi, Metropolis, and others at LANL [30] to perform shielding and dose calculations, but was extended to become a powerful tool to simulate nuclear reactor, and weapon systems. Chapter one of the MCNP5 Users Manual defines the capabilities of the software:

> MCNP is a general-purpose, continuous-energy, generalized-geometry, time-dependent, coupled neutron/photon/electron Monte Carlo transport code. It can be used in several transport modes: neutron only, photon only, electron only, combined neutron/photon transport where the photons are produced by neutron interactions, neutron/photon/electron, photon/electron, or electron/photon. The neutron energy regime is from $10-^{11}$ MeV to 20 MeV for all isotopes and up to 150 MeV for some isotopes, the photon energy regime is from 1 keV to 100 GeV, and the electron energy regime is from 1 KeV to 1 GeV. The capability to calculate keff eigenvalues for fissile systems is also a standard feature. [4]

The basic procedure for running MCNP5 begins with the creation of an input file in which the user defines a system geometry, specifies materials, identifies neutron, photon, or electron sources, indicates the information or calculations that are desired, and sets limits on the number of iterations or how long to run the simulation.

The basic functioning of MCNP5 is to track an initial set number of particles (i.e., neutrons) and, using probability distributions or cross-sections, determining the results of collisions and interactions between those particles and the materials in the system. The software then keeps track of the particles and calculations that are relevant to obtain the desired solutions.

In order to model a critical reactor system and mathematically track the path of a particle in the system, MCNP5 uses the Boltzmann transport equation:

$$\Psi(r,v) = \int [\int \Psi(r',v') C(v' \to v, r') dv' + Q(r',v)] T(r' \to r, v) dr'$$

where:

$$\Psi(r,v) = \text{ particle collision density}$$

$$Q(r',v) = \text{ Source term}$$

$$C\left(v' \to v, r\right) = \text{ collision kernel, change velosity at fixed position}$$

$$T\left(r' \to r, v\right) = \text{transport kernel, change velosity at fixed velosity}$$

Equation 3: Boltzmann transport equation. From [30]

The Boltzmann transport equation is used to determine when collisions happen between the particles and the atoms around them, with the simplifying assumptions of a static, homogeneous medium, no time dependence, a Markovian system, where the next event is only determined by the previous event, no particle-particle interactions, no relativistic effects, no long-range forces, and no change in the medium due to particle interactions [30]. This leads to a random walk path, a path that involves moving a distance in a straight line, having an interaction, and then moving again. Each particle being tracked will have a random walk path the physics is in the calculations to determine the result of each interaction [30]. In order to determine first if there was an interaction, MCNP5 must access a large cross-section database, included with the code. The code looks up the cross-section of the particle at the relevant energy with the particular isotope it is interacting with. Then the code determines the type of interaction by weighing the probabilities of each possible interaction, absorption, elastic, inelastic, fission, etc., and randomly selecting one based on the probabilities. If the particle continues on after the interaction, or a new particle is created, the direction and speed will also be randomly determined based upon probability, or determined by kinematics, if applicable [30]. A tracked particle begins at a point source or an interaction that creates a new particle, and ends when the particle leaves the system or is consumed by an interaction. MCNP tracks many particles simultaneously in the described method; in order to gain meaningful information the code saves particle histories and tallies particular events. For a criticality

calculation, such as the one used in this research, the number of neutrons at the beginning and end of each cycle are tallied and divided to give a criticality value.

MCNP5 furthermore has the ability to choose which particles to track where and can weight which type of interaction the simulation will produce during an interaction, and what information is to be tallied and calculated. The MCNP5 software package gives the user access to vast amounts of information about the performance of a real world system because it simulates a real world system and given enough computational resources, will provide a statistically accurate result.

In the specific application of MCNP used in this thesis to calculate the criticality of a simplified LFR reactor system, the Monte Carlo method is applied to the an initial set of neutrons which interact with the atoms in the reactor (fuel as well as coolant) to probabilistically determine the criticality, or k-value of the specified geometrical arrangement of fuel and coolant mixes of various isotopes. Each introduced neutron is tracked until it collides with an atom of the reactor model at which point the code looks up the cross-sections for different types of interactions with the atom and then determines probabilistically what type of collision occurred and what the result of that collision is. Then the code continues to track the neutron or other by-products of the collision/interaction which could be a scatter, an absorption, a fission, or any one of several other possible interactions. In order to calculate the criticality of the system the code takes the number of neutrons tracked at the beginning of the cycle, adds the number of neutrons created as the result of fission, subtracts the number of neutrons lost through absorption or leakage across the boundaries of the system, and then it divides this result at the end of a given cycle number by the initial neutron count at the beginning of the cycle. Increased accuracy is obtained by increasing the number of neutrons actively tracked and by tracking them for greater numbers of cycles, effectively increasing the 'real-time' during which data is recorded. Increases in accuracy come at the expense of increases in the number of computations required.

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IV. PROCEDURE AND RESULTS

The major objectives of this thesis include the assessment of isotopic variability of lead in the environment to characterize the most favorable natural mixes from the standpoint of nuclear properties relevant to use as a reactor coolant; and assessment of the potential impact such a favorable mix could have on key parameters of a model lead cooled reactor. This chapter presents the procedure and results of the research in this thesis to achieve these objectives.

A. WHAT ISOTOPE MIXES OF LEAD ARE AVAILABLE IN NATURE

Based upon the known cross-sections of stable lead isotopes and thus concluding that only ^{208}Pb and ^{207}Pb enrichment can lead to improved flexibility in liquid lead reactor design, it is necessary to find natural sources of lead that are elevated in ^{208}Pb and/or ^{207}Pb.

1. Lead Mines

The first place to investigate to locate favorable isotopic mixes is lead mines. According to a 2011 United States Geological Survey the lead production breakdown by country is listed in Table 2.

Lead Production and Reserves		
Country	Production	Reserves
USA	400	7000
Australia	620	27000
Bolivia	90	1600
Canada	65	650
China	1600	13000
India	95	2600
Ireland	45	5600
Mexico	185	6000
Peru	280	600
Poland	35	1500
Russia	90	9200
South Africa	50	300
Sweden	65	1100
Other	330	4000
Total	4,100	80,000
Data is in thousands of metric tons. Data		
From USGS Mineral Commodity		
Summary, January 2011.		

Table 2.   List of largest lead producing countries. From [29].

Data on isotopic concentration of lead from lead mines in China, Australia, Canada, Mexico, Spain, and the United States were reproduced to evaluate the variation that is found among naturally occurring galena, the mineral from which most lead is mined.

Mine Location	^{204}Pb	^{206}Pb	^{207}Pb	^{208}Pb
Bathhurst, New Brunswick	1.370	24.950	21.530	52.140
Chicobi Lake, Quebec	1.608	21.530	23.460	53.400
San Antonio Mine, Mexico	1.343	25.280	21.210	52.160
New Dodgeville, Wisconsin	1.185	28.390	19.220	51.200
Joplin, Missouri	1.245	27.290	20.010	51.450
Ace Mine, Saskatchewan [15]	1.044	41.870	19.450	37.640
Nuevo Larado, Mexico	0.649	32.626	22.620	44.105
Forest City, Iowa	1.329	25.601	21.190	51.880
Modoc, Kansas	1.343	26.165	21.169	51.323
Henbury, Australia	1.981	18.922	20.567	58.530
Canon Diablo, Arizona [15]	1.990	18.830	20.581	58.599
China 1	1.340	25.292	21.014	52.355
China 2 [31]	1.340	25.263	21.003	52.394
Val d'Aran mining district, Spain	~ 1	24.901	21.540	52.559
Osor mining district, Spain [32]	~ 1	25.158	21.213	52.629

Table 3.   Isotopic concentrations of various lead mines by percent.

The isotopic concentrations are fairly constant across the different locations with average values of 51.5% ^{208}Pb, 21.1% ^{207}Pb, 26.1% ^{206}Pb, and 1.3% ^{204}Pb, which are all consistent with the recorded averages. The Henbury and Canon Diablo sources do have a slightly (~7%) higher ^{208}Pb concentration that will be evaluated in subsequent reactor simulations.

If one is to search for favorable sources of lead in mining deposits, consider the first six mines in Table 3; the first five of these were all taken from lead mines, while the sixth sample, Ace Mine, was taken from a uranium mine and is much higher in ^{206}Pb, and much lower in ^{208}Pb than the other five. This is a result of the ^{238}U decaying into ^{206}Pb, and the relative lack of thorium in the ore to produce ^{208}Pb. This example demonstrates that a thorium source would be the ideal place to find a high ^{208}Pb concentration lead. It is also demonstrated via lead-lead dating conducted by R. D. Russell that in order to assist in locating a mine with a high ^{207}Pb / ^{206}Pb, which would be favorable so long as it was

not at the expense of a lower ^{208}Pb content, one should find the oldest lead, as the ^{207}Pb / ^{206}Pb ratio grows over time [15]. Unfortunately it is also known by lead-lead dating that the oldest lead will contain the lowest ^{208}Pb / ^{204}Pb ratio [15], thus only the thorium to uranium ratio will be beneficial in estimating the likelihood of a specific lead source being favorable.

2.   Monazite Ores

Having seen that, besides thorium vs. uranium content, there are no good indicators that a lead mine will have favorable lead compositions for the desired nuclear reactor application, it is appropriate to seek other places from which to potentially extract lead. Because ^{208}Pb is made by the decay of thorium, a favorable lead could potentially be extracted from a thorium ore. The most common thorium ore is monazite, which is a sand material from which various minerals are extracted. A study of various monazite ores that was recently complete [33] included U-Th-Pb dating information, which is reproduced in Table 4.

	ppm of		Age in million years based on		
Monazite	U	Th	^{207}Pb/^{235}U	^{206}Pb/^{238}U	^{208}Pb/^{232}Th
16-F-6	2392	42065	2832	2845	2957
Elk	2026	126033	1458	1504	1442
Namaqua-1	1291	82676	1050	1047	1061
Iveland	759	36733	941	934	929
E0013	5385	77177	590	591	590
Manangoutry	1098	116988	568	559	563
M1	2865	104699	524	525	535
M2	3440	132961	521	523	528
M3	2946	80980	521	521	534
M4	2882	77169	521	519	525
INDIA	2087	41372	510	514	531
Jefferson	4923	51288	371	365	369
Maine1	5350	141043	277	275	289
Maine2	3939	111984	280	283	304

Table 4.   U-Th-Pb ages of the standard monazites. From [33].

Using this data, the equations for radiological dating, the assumption that 1.4% of lead is ^{204}Pb, that the number of daughter isotopes was initially zero, and that the ^{238}U / ^{235}U ratio was 99.3% and .7% the isotopic composition of the lead in the monazite samples were calculated in Table 5.

Monazite	^{204}Pb	^{206}Pb	^{207}Pb	^{208}Pb
16-F-6	1.400	13.446	17.731	67.423
Elk	1.400	4.619	12.775	81.206
Namaqua-1	1.400	3.980	16.398	78.223
Iveland	1.400	4.851	22.625	71.124
E0013	1.400	7.588	57.320	33.692
Manangoutry	1.400	2.327	18.644	77.630
M1	1.400	4.662	39.855	54.084
M2	1.400	4.536	38.929	55.136
M3	1.400	5.359	46.186	47.055
M4	1.400	5.450	47.153	45.997
INDIA	1.400	6.114	53.445	39.041
Jefferson	1.400	5.883	73.248	19.468
Maine1	1.400	3.736	62.175	32.688
Maine2	1.400	3.679	59.453	35.469

Table 5.   Isotopic concentrations of various monazite ores by percent.

The isotopic breakdown of lead from monazite ores is promising due to the very high ^{208}Pb values in some samples, leading to an average of 52.73%, also the ^{207}Pb concentration is quite high at 42.4%, and the ^{206}Pb concentration which is low at only 5.4%. The best of these samples will be included in reactor simulations to determine how favorable they are for liquid lead reactor coolant use.

	ppm of		
Monazite	Th	Pb	Pb / Th
16-F-6	42065	7067	0.168
Elk	126033	6630	0.053
Namaqua-1	82676	3076	0.037
Iveland	36733	1193	0.032
E0013	77177	1926	0.025
Manangoutry	116988	2520	0.022
M1	104699	2283	0.022
M2	132961	2841	0.021
M3	80980	1811	0.022
M4	77169	1702	0.022
INDIA	41372	977	0.024
Jefferson	51288	867	0.017
Maine1	141043	1773	0.013
Maine2	111984	1438	0.013

Table 6. Concentration of thorium and lead in monazite ores. From [33].

Monazite is currently mined for thorium economically and has a lead content between one and seventeen percent, as much lead as it has thorium, as seen in Table 6. Thus lead could potentially be extracted from the waste products of thorium mining.

3. Rare Earth Metal Ores

Thorium is known to be geochemically similar to rare earth metals [28]; as such it is often found in minerals that are mined to extract rare earth metals.

Ore	Chemical Composition
Allanite	$(Ca,Ce,\textbf{Th})_2(Al,Fe,Mg)_3Si_3O_{12}(OH)$
Monazite	$(Ce,La,Pr,Nd,\textbf{Th},Y)PO_4$
Parisite	$2(Ce,La,Di,\textbf{Th})OF.CaO.3CO_3$
Polymignite	$(Ca,Fe,Y,\textbf{Th})(Nb,Ti,Ta,Zr)O_4$
Euxenite	$(Y,Ca,Er,La,Ce,\textbf{U},\textbf{Th})(Nb,Ta,Ti)_2O_6$
Cheralite	$(Ca,Ce,\textbf{Th})(P,Si)O_4$
Samarskite	$(Y,Er,Ce,\textbf{U},Ca,Fe,Pb,\textbf{Th})(Nb,Ta,Ti,Sn)_2O_6$
Thorogummite	$(\textbf{Th}(SiO4)_{1-X}(OH)_{4X})$
Davidite	$(La,Ce)(Y,\textbf{U},Fe^{+2})(Ti,Fe^{3+})_{20}(O,OH)_{38}$
Fergusonite	$(Y,Er,Ce,Fe)(Nb,Ta,Ti)O_4$
Loparite	$(Ce,Na,Ca)(Ti,Nb)O_3$
Bastnäsite	$(Ce,La,Di)F.CO_2$

Table 7.   Chemical composition of some rare earth ores with Th and U occurrence.
From [34].

It can be seen in Table 7 that of the twelve rare earth ores, eight have thorium in them and of those eight, only two have uranium also. It is therefore reasonable to anticipate that the remaining six ores potentially have a high [208]Pb concentration. Though the actual lead concentrations are not available, it can further be assumed that the [208]Pb would be highest in the older ores. Favorable lead could theoretically be extracted from the waste of rare earth ore processing. Due to the uncertainties in the lead composition of such ores and the very limited available data on lead isotopic composition of rare-earth ores, lead as a byproduct of rare earth mining was not included in the simulations.

B.   SIMULATED REACTOR MODEL

In order to address the question, "what benefit in the design of a lead-cooled reactor can be expected from the use of favorable isotopic mixes of lead?," a representative model of the reactor system must first be built. The model that was constructed was a highly simplified reactor based upon parameters defined in the SSTAR concept (see Figure 10). The model reactor vessel is a cylindrical structure that is 15 meters in height, 3 meters in diameter, and consists solely of lead coolant. The core region of the reactor is modeled as a smaller cylinder of .8 meter height and 1 meter diameter centered within the reactor vessel and consisting of half fuel and half lead

coolant. This basic simplified model of a reactor was analyzed with various lead isotope mixes as the coolant within the core region and throughout the vessel, while tuning the fuel enrichment to reach a criticality of one. The actual SSTAR reactor has many other components and complexities; however the relevant results are the different critical fuel enrichments corresponding to the various isotopic mixes of lead, not the required amount of enrichment for the actual system. Due to the fact that this difference provides the key indication of the impact of a particular coolant composition, the simplified reactor model is believed to be sufficient to indicate the potential of favorable isotopic mixes of lead coolant.

C.   ISOTOPIC MIXES TO TEST

Various lead coolants were simulated in exercising the above the model. The first set of simulations were run assuming that the lead coolant consisted of an isotopically pure mass of each of the stable isotopes of lead: ^{204}Pb, ^{206}Pb, ^{207}Pb, and ^{208}Pb. The first four single isotope coolants were run in order to establish benchmarks of what kind of effect the isotopic content of lead coolant has on a meaningful parameter that is a primary indicator of importance in reactor design. The measurable value selected for all the simulations is the required enrichment for criticality. This is a good benchmark when using MCNP as it allows for a minimal change in code in order to complete each simulation run. It should be noted that, assuming favorable lead isotope mixes have significant impacts on the required enrichment, designers and engineering teams could take alternative options to improve the reactor design such as modifying the reactor dimensions (e.g., reducing the size of the core or the entire module) or using a lower enrichment, and therefore less expensive, nuclear fuel.

Following the simulations based on single pure isotopes of lead, a second baseline simulation was carried out by assuming the lead coolant was based on the published natural isotopic mix of lead, This baseline simulation is important in establishing the nominal criticality characteristics of the model reactor from which to judge the potential improvements that might be achieved with a more favorable isotopic mix. The final

simulations were based on the anticipated favorable isotopic mixes that can be expected to be found in natural sources, as revealed by the lead isotope geochemistry study in this thesis.

D. MCNP5 CODE INPUTS AND SIMULATIONS FOR THE SIMPLIFIED REACTOR MODEL

The code inputs for the simplified model provide a simple approximation of a cylindrical reactor measuring fifteen meters in height and three meters in diameter with a second cylinder centered inside the first that was eighty centimeters in height and one meter in diameter. The larger cylinder representing the reactor vessel and its contained coolant is composed of pure lead with various isotopic concentrations, at a density of 10.66 grams per a cubic centimeter, the density of molten natural lead at its melting point (no adjustment being made for possible isotopic differences in density which would, in any case, be quite small). The second cylinder representing the reactor core is composed of fifty percent lead coolant, and fifty percent uranium-nitride (U_2N_3) fuel, with a density of 10.96 grams per a cubic centimeter, representing the average of the uranium-nitride density (11.3 grams per a cubic centimeter) and lead. No simulations of LBE were tested due to the corrosive nature of LBE, as well as its previously proven success.

The simulations were run for 250 cycles tracking 5000 neutrons in each cycle. Each simulation began with a fission start point in the center of the system. Because of the anomalous effect on the system criticality of the central introduction of the initial neutron, the first fifty cycles were ignored and the code began tracking and averaging the results during the remaining 50 cycles to determine criticality. The code also calculated a standard deviation for the criticality value of the cycles counted. The simulations were run with an initial ratio of U_{235} to total Uranium of fifteen percent and the ratio was then adjusted in order to achieve a criticality value of one. One run each was performed for a pure ^{204}Pb, ^{206}Pb, ^{207}Pb, ^{208}Pb, another run for natural lead, and a run was done with a mix of the normal concentration (52.4 %) of ^{208}Pb and the remaining lead (47.6 %) being ^{207}Pb. The variations in the enrichment values demonstrate the advantage of using various lead isotopic mixes for the coolant.

41

Figure 11 presents the input file for the model calculation using natural lead as the coolant. Input files for the other simulations are similar.

```
      SSTAR lead isotope variations
C     Cell Cards
10    120 -10.66 -1 2 -3 (4:-5:6) imp:n=1 $ coolant cylinder
20    121 -10.98 -4 5 -6          imp:n=1 $ fuel cylinder
30    0 1:3:-2                     imp:n=0 $ surrounding void

C     Surface Cards
1     cz  150     $ coolant cylinder 1.5 meter radius
2     pz  0       $ coolant base at 0 meters
3     pz  1500    $ coolant top at 15 meters
4     cz  50      $ fuel cylinder .5 meter radius
5     pz  710     $ fuel base at 7.1 meters
6     pz  790     $ fuel top at 7.9 meters

C     Data Cards
C     Material Data Cards
m10   82204.70c 1.0     $ lead 204
m11   82204.70c .50
      92235.66c .04917
      92238.66c .15083
      7015.66c  .30     $ lead 204 fuel mix
m20   82206.66c 1.0     $ lead 206
m21   82206.66c .50
      92235.66c .0424
      92238.66c .1576
      7015.66c  .30     $ lead 206 fuel mix
m30   82207.66c 1.0     $ lead 207
m31   82207.66c .50
      92235.66c .0372
      92238.66c .1628
      7015.66c  .30     $ lead 207 fuel mix
m40   82208.66c 1.0     $ lead 208
m41   82208.66c .50
      92235.66c .03375
      92238.66c .16625
      7015.66c  .30     $ lead 208 fuel mix
m50   82204.70c .014
      82206.66c .241
      82207.66c .221
      82208.66c .524    $ natural lead
m51   82204.70c .0070
      82206.66c .1205
      82207.66c .1105
      82208.66c .2620
      92235.66c .0367
      92238.66c .1633
      7015.66c  .30     $ natural lead fuel mix
m60   82207.66c .476
      82208.66c .524    $ lead 207 enriched
m61   82207.66c .238
      82208.66c .2620
      92235.66c .03547
      92238.66c .16453
      7015.66c  .30     $ lead 207 enriched fuel mix
m70   82204.70c .014
      82206.66c .0462
      82207.66c .1278
      82208.66c .8120   $ elk lead
```

Figure 11.   MCNP5 code used for benchmark simulations

```
m71  82204.70c .0070
     82206.66c .0231
     82207.66c .0639
     82208.66c .4060
     92235.66c .03555
     92238.66c .16445
     7015.66c  .30      $ elk lead fuel mix
m80  82204.70c .014
     82206.66c .0398
     82207.66c .1640
     82208.66c .7822    $ namaqua-1 lead
m81  82204.70c .0070
     82206.66c .0199
     82207.66c .0820
     82208.66c .3911
     92235.66c .03565
     92238.66c .16435
     7015.66c  .30      $ namaqua-1 lead fuel mix
m90  82204.70c .014
     82206.66c .0588
     82207.66c .7325
     82208.66c .1947    $ jefferson lead
m91  82204.70c .0070
     82206.66c .0294
     82207.66c .36625
     82208.66c .09735
     92235.66c .03705
     92238.66c .16295
     7015.66c  .30      $ jefferson lead fuel mix
m100 82204.70c .014
     82206.66c .0374
     82207.66c .6217
     82208.66c .3269    $ maine1 lead
m101 82204.70c .0070
     82206.66c .0187
     82207.66c .31085
     82208.66c .16345
     92235.66c .03662
     92238.66c .16338
     7015.66c  .30      $ maine1 lead fuel mix
m110 82204.70c .0198
     82206.66c .1892
     82207.66c .2057
     82208.66c .5853    $ henbury lead
m111 82204.70c .0099
     82206.66c .0946
     82207.66c .10285
     82208.66c .29265
     92235.66c .0366
     92238.66c .1634
     7015.66c  .30      $ henbury lead fuel mix
m120 82204.70c .0199
     82206.66c .1883
     82207.66c .2058
     82208.66c .586     $ canyon diablo lead
m121 82204.70c .00995
     82206.66c .09415
     82207.66c .1029
     82208.66c .293
     92235.66c .0366
     92238.66c .1634
     7015.66c  .30      $ canyon diablo lead fuel mix
C    Criticality Control Cards
kcode 5000 1.0 50 250
ksrc                            0              0              750
```

Figure 11. (continued) MCNP5 code used for benchmark simulations

The MCNP5 input file consists of four sections as shown in Figure 11. The first section, the cell cards, defines the separate cells, their material, density, and geometric boundaries. The second section, the surface cards, lists the surfaces that are used in creating the cells. The third section, the material cards, lists the materials that are used to fill the cells. The example in Figure 11 has many unused materials because each isotopic mix of lead is included but only one is used each run. The final section, the criticality control cards, are used to define what particles are to be tracked, how many of them, how many runs to perform, how many to ignore, and where the fission start point is.

In the simulations run for this thesis, the value of the ^{235}U enrichment was started at fifteen percent and was raised incrementally until a keff value near 1.0 with a three decimal point accuracy was achieved.

E. RESULTS OF BENCHMARK SIMULATIONS

The benchmark simulations provided the following results:

	k_{eff}	^{235}U Enrichment	Standard deviation
^{204}Pb	1.00040	24.585 %	0.00050
^{206}Pb	1.00030	21.200 %	0.00060
^{207}Pb	0.99970	18.600 %	0.00050
^{208}Pb	0.99992	16.875 %	0.00059
Natural Pb	0.99992	18.350 %	0.00052

Table 8.   Required enrichment for criticality ($k_{eff} \sim 1$)-1.

The data from the Table 8 benchmark results indicates that only the pure isotope ^{208}Pb would provide an advantage over natural lead terms of offering a reduction in the fuel enrichment required for criticality. Thus enrichment in ^{208}Pb (or possibly enrichment in ^{207}Pb so long as ^{208}Pb does not decrease and the other two isotopes are simply displaced) will lead to a benefit over natural lead. The effect of the presence of ^{204}Pb is considered minimal due to its low, 1.4%, concentration in natural lead; therefore, small variations in ^{204}Pb can generally be ignored. To determine the usefulness of ^{207}Pb enrichment, a simulation based on a 52.4% ^{208}Pb and 47.6% ^{207}Pb mix was run (Table 9).

	k_{eff}	Enrichment	Standard deviation
47.6% ^{207}Pb	1.00016	17.735 %	0.00051

Table 9.   Required enrichment for criticality ($k_{eff} \sim 1$)-2.

The run in Table 9 indicates that a coolant enriched in ^{207}Pb could be viable as well; however this test assumes that the ^{207}Pb content is increased by displacing the ^{204}Pb and ^{206}Pb isotopes only. It can be concluded that a lead source will only be an improvement if it has a small, (i.e., lower than 2% concentration of ^{204}Pb), a ^{207}Pb concentration below 48%, and a concentration of ^{208}Pb greater than 52%.

The goal of this investigation of various lead sources is to find the one which will provide the most favorable isotopic mix of lead that is available in nature for use as coolant in a reactor. Ideally the best source would be one of pure ^{208}Pb, however it is unlikely that pure ^{208}Pb will be found in nature, thus using the guidelines establishing by the benchmark tests a source of favorable lead can be identified if found.

F.   ADDITIONAL MCNP RESULTS

Having seen the variations in isotopic concentration of various potential sources of lead, the necessary fuel enrichment was next determined for promising samples using the same simulated reactor that was used earlier for the benchmark tests; however for the final run of tests the number of neutrons tracked was increased to 10,000 and the number of cycles run was increased to 1000. The samples selected for final runs are the two monazite samples highest in ^{208}Pb, the two monazite samples highest in ^{207}Pb, and the 2 lead mine samples highest in ^{208}Pb. The results from an increased accuracy run of the benchmark tests are also included in Table 10.

Lead source	Isotopic concentrations				k_{eff}	^{235}U Enrichment	Standard deviation
	^{204}Pb	^{206}Pb	^{207}Pb	^{208}Pb			
Benchmark simulations							
^{204}Pb	100	0	0	0.00	1.00040	24.59%	0.00050
^{206}Pb	0	100	0	0.00	1.00030	21.20%	0.00060
^{207}Pb	0	0	100	0.00	0.99970	18.60%	0.00050
^{208}Pb	0	0	0	100.00	0.99992	16.88%	0.00059
Natural Pb	1.4	24.1	22.1	52.40	0.99992	18.35%	0.00052
Monazite high ^{208}Pb							
Elk	1.40	4.62	12.78	81.20	0.99999	17.78%	0.00046
Namaqua-1	1.40	3.98	16.40	78.22	0.99983	17.83%	0.00053
Monazite high ^{207}Pb							
Jefferson	1.40	5.88	73.25	19.47	1.00049	18.53%	0.00051
Maine1	1.40	3.74	62.17	32.69	1.00011	18.31%	0.00057
Lead mines high ^{208}Pb							
Henbury	1.98	18.92	20.57	58.53	1.00039	18.30%	0.00051
Canon Diablo	1.99	18.83	20.58	58.60	0.99982	18.30%	0.00054

Table 10.   Required enrichment for SSTAR based model reactor to achieve criticality using various natural lead concentrations.

V.   ANALYSIS AND CONCLUSION

Having investigated the nuclear properties of lead, and determining the isotopic content of various favorable alternative lead sources, the practical results of this data are discussed in this chapter.

A.   EXTRACTION OF LEAD FROM ALTERNATIVE SOURCES

It was cited earlier that artificial ^{208}Pb enrichment is prohibitively expensive with estimated costs of "1000–2000 U.S. $/kg." [13] In order to utilize alternative natural sources such as monazite, a method to obtain lead as a byproduct of monazite processing for thorium and rare earth metals extraction would need to be developed. The residual waste products from current ore processing include lead and other metals such as aluminum and iron. This would have to separated and extracted through a process that has not been developed. The process, however, would likely be much simpler and less expensive than lead enrichment because the lead can be chemically separated from the ore residue in contrast to separating the chemically-identical isotopes of lead in isotope enrichment.

The actual mining costs would be relatively small because monazite and rare earth metals are already mined economically for thorium and rare earth, thus all that would be required is another step to be added to the processing of the ore, that separates the lead.
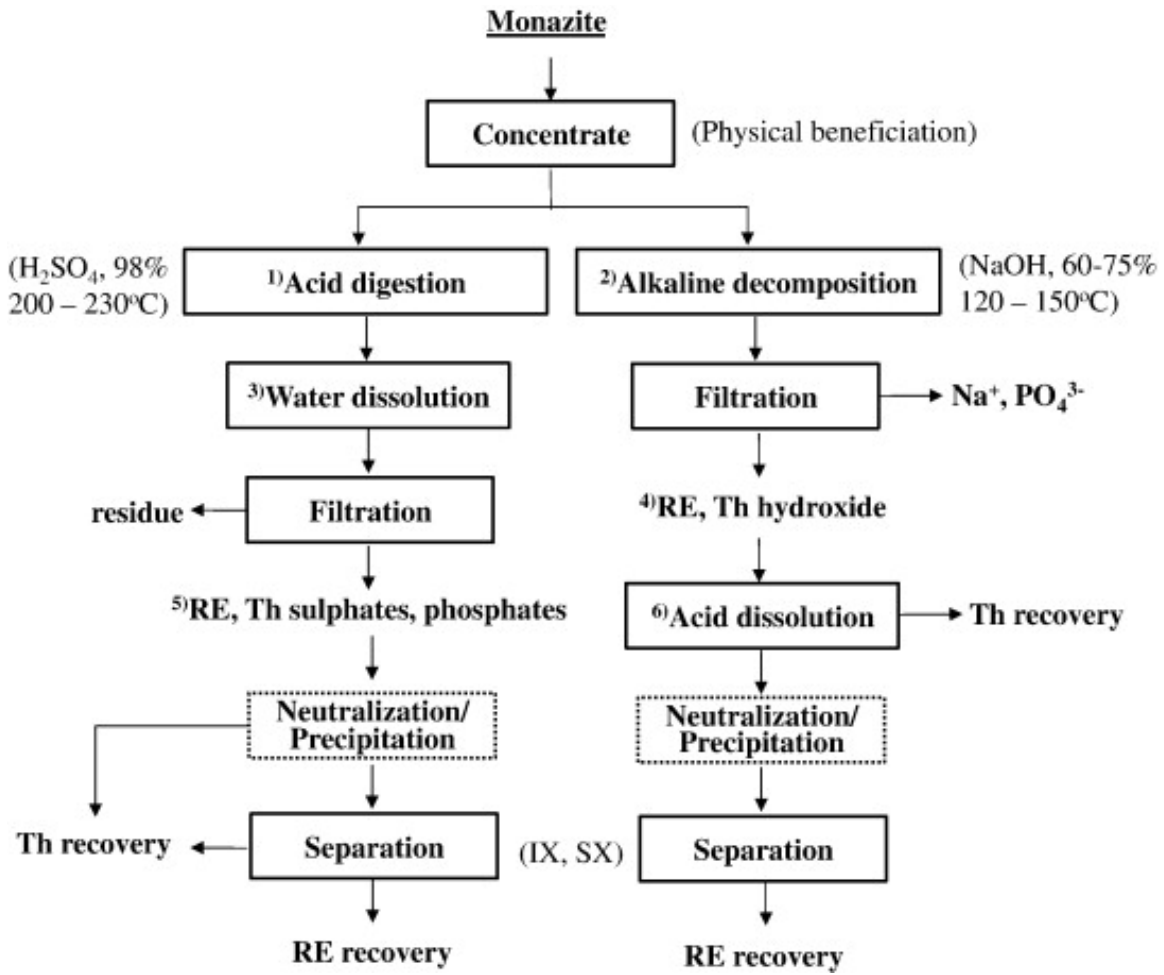
Figure 12.   Flowchart of monazite processing methods. From [35].

Though there are various monazite ore processing methods all ore processing produces waste and it is out of this waste from which the lead must be pulled as seen in Figure 12.

A study of the content of the various waste created during the ore processing would have to be conducted as well as a cost estimation of the development of a lead extraction process either from the waste, or an existing step in the current process.

B. DESIGN IMPLICATIONS OF ALTERNATIVE LEAD SOURCES

It has been shown that for the simplified model of SSTAR that was simulated, the required fuel enrichment can be reduced by about one percent by utilizing lead extracted from monazite as the coolant. The fuel enrichment required for criticality is only one potential design implication for lead cooled reactors. The accepted safe maximum enrichment for commercial reactors, for nuclear proliferation reasons is 20% [25]. As such rather than changing the ^{235}U enrichment, the size of the core could be made smaller, or the reactor-vessel diameter could be made slightly smaller because of the greater neutron reflection of the ^{208}Pb. This improved lead allows a level of flexibility for the design process which could lead to a more transportable SSTAR, or other small LFR, with no loss in power output.

The design flexibility could lead to a wider application of the SSTAR, beyond the localization of the power grid and the leasing to resource poor areas; for example the greater portability could allow for transportable energy sources for use by military and relief forces that have lower fuel requirements, and a corresponding lowering of the required logistics support. Also the Navy could utilize models of small reactors as drop-in propulsion sources rather than having the reactors built into their platforms.

C. CONCLUSION

The one percent lowering of ^{235}U may seem fairly small; however using the standard of 20 percent maximum uranium enrichment, the comparison of how thick the lead encasement need be for criticality demonstrates the potential for minimization of liquid lead reactors via favorable lead source utilization.

	k_{eff}	Standard deviation	Case Diameter
Natural Pb	1.00035	.00057	1.820 meters
Elk	.99984	.00064	1.728 meters

Table 11.   Outer case diameter for natural lead and monazite lead

The results in Table 11 demonstrate the potential of an improved lead source to reduce the diameter of an SSTAR or similar unit by up to 5%. Before any benefits can be realized from improved lead sources, more research has to be done to determine the feasibility, and economics of monazite ore lead extraction. Furthermore the design effects must be determined on more complex reactor models, and simulations. This research has shown that the most promising improvements in lead isotopic mix for lead-cooled reactor designs are those that are elevated in ^{208}Pb.

VI.   SUMMARY OF RESEARCH AND FUTURE WORK

The goal of this thesis was to answer the questions: (1) what naturally-occurring favorable mixes of lead are available? and (2) what level of design benefit would the use of such favorable isotopic mixes of lead provide? In order to answer the questions a geochemical analysis was conducted to find favorable naturally occurring lead. The favorable lead was simulated using MCNP5 in a simplified model of the SSTAR reactor, and the results were measured by the required enrichment in order to achieve a criticality of 1.

Lead is primarily made up of the radiogenic isotopes that are the end products of uranium and thorium decay; as such it can occur in different isotopic mixes due to the various geochemical processes that affect uranium and thorium. Most natural occurrences of lead are close to the overall average natural abundance; however, lead extracted from thorium ores can be significantly higher in the ^{208}Pb isotope, which is preferable for use in a lead cooled fast reactor due to its reduced tendency to parasitically absorb neutrons. In particular monazite ore is a promising source of high ^{208}Pb lead. Such a source could be exploited to produce lead for use in lead cooled fast reactors.

In order to investigate the potential implications for nuclear reactor designs, and to assist in the determination of lead isotopic content in various lead sources, an understanding of key nuclear physics concepts was required. The Bateman equations, radioactive dating, nuclear cross-sections, and reactor criticality were explored, and consequently utilized in calculations done both to determine lead isotopic mixes, and by MCNP code to simulate a reactor.

SSTAR is a Generation IV reactor design that uses lead as a primary coolant. The design is based on many years of experience with other reactors and was created to be a transportable reactor, with autonomous operation. It would give the option of nuclear power to many developing countries. The reactor has a fairly straightforward geometry that allowed for a simple model to be constructed in order to run simulations to determine the effect of isotopic concentration of lead on the performance of the reactor.

A Monte Carlo particle transport code, called MCNP5, was used to simulate the SSTAR and run tests with various lead isotope mixes. It was determined that lead extracted from particular monazite ores would be the best for use in SSTAR. The ^{208}Pb enriched lead would allow for efficiency increases that could be translated to a smaller overall design, lower core enrichment, or greater power output from a design of the same size.

The reactor simulations determined that up to a one percent decrease in reactor core enrichment could be achieved in the simplified rector modeled for this work. However the actual SSTAR reactor core is smaller than the one modeled which could lead to a greater percent reduction in required enrichment for a k_{eff} of 1.

A.   FUTURE WORK

In order to implement this research, additional follow-on work can be recommended. Specifically future projects could include:

A further analysis of monazite ores for lead isotopic content.

A feasibility and cost analysis of extracting lead from monazite ore.

A study of lead isotopic content in rare earth ores.

A precise model of SSTAR with varying lead isotopic content in the primary coolant circuit.

Any of the projects above could further the objectives of this thesis and lead to advancement in the capabilities of the SSTAR project.

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